



Development and evaluation of the unified tropospheric–stratospheric chemistry extension (UCX) for the global chemistry–transport model GEOS-Chem



Sebastian D. Eastham^a, Debra K. Weisenstein^b, Steven R.H. Barrett^{a,*}

^a *Laboratory for Aviation and the Environment, Department of Aeronautics and Astronautics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA*

^b *Harvard School of Engineering and Applied Sciences, 29 Oxford Street, Cambridge, MA 02138, USA*

HIGHLIGHTS

- We extend GEOS-Chem with a unified tropospheric–stratospheric chemical mechanism.
- Emission, transport, mixed-phase chemistry, photolysis and destruction of new species.
- Fast-JX couples online stratospheric aerosols to tropospheric photochemistry.
- Models 90% of maximum ozone depletion in 2006 Antarctic ozone hole.
- Widely-used model now captures coupled stratospheric–tropospheric responses.

ARTICLE INFO

Article history:

Received 25 September 2013

Received in revised form

30 January 2014

Accepted 4 February 2014

Available online 6 February 2014

Keywords:

GEOS-Chem

Stratospheric chemistry

Ozone hole

Model development

Model validation

ABSTRACT

Global chemistry–transport models (CTMs) typically use simplified parameterizations or relaxation to climatology to estimate the chemical behavior of the stratosphere only in the context of its impact on tropospheric chemistry. This limits investigation of stratospheric chemistry and interactions between tropospheric and stratospheric chemistry–transport processes. We incorporate stratospheric chemical and physical processes into the model GEOS-Chem in the form of a unified chemistry extension (UCX). The stratospheric chemistry framework from NASA's Global Modeling Initiative (GMI) is updated in accordance with JPL 10-06 and combined with GEOS-Chem's existing widely applied and validated tropospheric chemistry to form a single, unified gas-phase chemistry scheme. Aerosol calculations are extended to include heterogeneous halogen chemistry and the formation, sedimentation and evaporation of polar stratospheric clouds (PSCs) as well as background liquid binary sulfate (LBS) aerosols. The Fast-JX v7.0a photolysis scheme replaces a hybrid of Fast-J and Fast-JX v6.2, allowing photolytic destruction at frequencies relevant to the stratosphere and of species not previously modeled. Finally, new boundary conditions are implemented to cover both surface emissions of new species and mesospheric behavior. Results for four simulation years (2004–2007) are compared to those from the original, tropospheric model and to in situ and satellite-based measurements. We use these comparisons to show that the extended model is capable of modeling stratospheric chemistry efficiently without compromising the accuracy of the model at lower altitudes, perturbing mean OH below 250 hPa by less than 5% while successfully capturing stratospheric behavior not previously captured in GEOS-Chem such as formation and collapse of the Antarctic ozone hole. These extensions (with supporting validation and intercomparison) enable an existing and extensively validated tropospheric CTM to be used to investigate a broader set of atmospheric chemistry problems and leverages GEOS-Chem's existing tropospheric treatment.

© 2014 Elsevier Ltd. All rights reserved.

1. Introduction

GEOS-Chem is a three-dimensional global tropospheric chemical–transport model (CTM) initially developed at Harvard University in the late 1990s (Bey et al., 2001) and collaboratively

* Corresponding author.

E-mail address: sbarrett@mit.edu (S.R.H. Barrett).

Table 1

Comparison of existing models to GEOS-Chem UCX. Models with distinct chemical mechanisms for the troposphere and stratosphere are shown here with two counts (tropospheric/stratospheric). Although GEOS-Chem can be run with secondary organic aerosols (SOA), these are not modeled in the default setup.

	Species	Kinetic reactions	Photolysis reactions	Soot	Organic carbon	Dust	Tropospheric sulfates	SOA	Sea salt	Stratospheric aerosols	Focus region
GC UCX	132	344	89	2	2	7	Y	Y/N	2	2	T, S
GEOS-Chem ^a	104	240	55	2	2	7	Y	Y/N	2	0	T
CAM-chem ^b	117	263	69	2	2	4	Y	Y	4	4	T
MOZART-3 ^c	108	236	71	2	2	0	Y	Y	4	4	T, S, M
MOZART-4 ^d	85	161	39	2	2	4	Y	Y	4	0	T
TM-5 ^e	42	64	16	0	0	0	Y	N	0	0	T, S
GMI ^f	46	116	38	1	0	0	N	N	0	4	T, S
OSLO CTM3 ^g	97	88/112	17/47	4	4	2	Y	Y	8	3	T, S
TOMCAT ^h	41	93	25	0	0	0	N	N	0	0	T
SLIMCAT ⁱ	43	109	29	0	0	0	N	N	0	3	S
CMAM ^j	44	93	34	0	0	0	N	N	0	1	S, M

^a Bey et al., 2001.

^b Lamarque et al., 2012.

^c Kinnison et al., 2007.

^d Emmons et al., 2010.

^e Huijnen et al., 2010.

^f Considine et al., 2000; Rotman and Tannahill, 2001.

^g Søvde et al., 2012.

^h Arnold et al., 2005.

ⁱ Chipperfield, 2006.

^j De Grandpré et al., 2000.

updated since (<http://www.geos-chem.org>). It has seen widespread use for investigations of the atmospheric response to emissions and natural phenomena (Barkley et al., 2011; Duncan Fairlie et al., 2007; Mao et al., 2010). The bulk of chemical reactions are accounted for only within the troposphere. Stratospheric chemistry is approximated through two primary mechanisms. The linearized ozone (Linoz) method (McLinden et al., 2000) is employed to estimate stratospheric ozone concentrations. Stratospheric bromine species concentrations are specified from climatology (Parrella et al., 2012). The evolution of most other species in the stratosphere is calculated based on archived monthly mean production rates and loss frequencies from NASA's Global Modeling Initiative (GMI) code (Murray et al., 2012; Rotman et al., 2001).

We develop the GEOS-Chem tropospheric–stratospheric unified chemistry extension (UCX) to include stratospheric chemistry in GEOS-Chem, thus enabling capture of stratospheric responses and troposphere–stratosphere coupling. Additionally, investigations of chemical feedbacks between stratospheric ozone and aerosols and tropospheric photochemistry are made possible through extension of photolysis to the stratopause, calculation of J-values for shorter wavelengths and improved modeling of high-altitude aerosols.

The paper is structured as follows: Section 2 outlines the changes and extensions which have been applied to GEOS-Chem with the aim of producing an accurate, unified model of the troposphere and stratosphere. Section 3 intercompares the results from the extended model against existing model results and validates against historical observations, with conclusions summarized in Section 4. The purpose of these extensions (with supporting validation and intercomparison) is to enable an existing and extensively validated community tropospheric CTM to be used to investigate a broader set of atmospheric chemistry problems, while leveraging the tropospheric chemistry of GEOS-Chem.

2. Model description

The base model is GEOS-Chem version 9-01-03. The extended (UCX) version models 132 chemical species, with long-lived chemical families transported in 54 tracers. For the simulations presented in this paper, we use GEOS-5 meteorological fields from NASA's Global Modeling and Assimilation Office (GMAO) over 72 hybrid sigma-eta pressure levels extending from the surface to

0.01 hPa. For computational efficiency we employ a $4^\circ \times 5^\circ$ horizontal resolution, although the model can be driven at higher resolutions including nested grid modeling at (for example) $0.5^\circ \times 0.667^\circ$ (Barkley et al., 2011). The UCX makes no changes to the global transport, convection or mixing processes.

Table 1 shows the relative scope of GEOS-Chem with and without the UCX, along with some other existing models. The “Focus Region” column corresponds to the part of the atmosphere–troposphere, stratosphere, mesosphere or a combination – is the primary focus of the model.

2.1. Chemistry

The UCX extends the chemistry mechanism to include reactions relevant to the stratosphere. As shown in Table 1, we add 28 species and 104 kinetic reactions, including 8 heterogeneous reactions, along with 34 photolytic decompositions. These were identified by comparing reactions and processes already present to those included in the GMI stratospheric chemistry mechanism (Rotman et al., 2001). Reaction constants were updated in line with JPL 10-06 (Sander et al., 2011). A full listing of modeled species and reactions is given in the Supplementary information (SI), available online.

2.1.1. Gas-phase kinetic chemistry

We explicitly model atomic oxygen [both $O(^3P)$ and $O(^1D)$]. The base model neglects them as intermediates due to their short lifetimes. Although these species still exhibit short lifetimes in the stratosphere, their importance in correctly modeling stratospheric chemistry result in the need for these reactions to be explicitly considered. We also explicitly model ground state atomic hydrogen (H) and nitrogen (N) as active species.

We also combine GEOS-Chem's bromine mechanism (Parrella et al., 2012) with an adapted version of GMI's chlorine mechanism. Chlorine is transported to the stratosphere in long-lived organic species such as CFCs and HCFCs, followed by the release of active chlorine and subsequent sequestration in reservoir species (ClONO₂ and HCl). Halon species H-1211, H-1301 and H-2402 are added as organic sources of bromine. Heterogeneous halogen chemistry is also modeled (see Section 2.1.3). Dry deposition and

Download English Version:

<https://daneshyari.com/en/article/6340133>

Download Persian Version:

<https://daneshyari.com/article/6340133>

[Daneshyari.com](https://daneshyari.com)